

NASA Technical Memorandum 85808

NASA-TM-85808 19840017365

MOLECULAR VELOCIMETRY USING STIMULATED RAMAN SPECTROSCOPY

REGINALD J. EXTON AND MERVIN E. HILLARD

MAY 1984

FOR REFERENCE

NOT TO BE TAKEN FROM THIS ROOM

LIBRARY COPY

JUN 1 1984

LANGLEY RESEARCH CENTER
LIBRARY, NASA
HAMPTON, VIRGINIA



National Aeronautics and
Space Administration

Langley Research Center
Hampton, Virginia 23665

DISPLAY 20/2/1

84N25433*# ISSUE 15 PAGE 2405 CATEGORY 72 RPT#: NASA-TM-85808 NAS
1.15:85808 84/05/00 23 PAGES UNCLASSIFIED DOCUMENT

UTTL: Molecular velocimetry using stimulated Raman spectroscopy

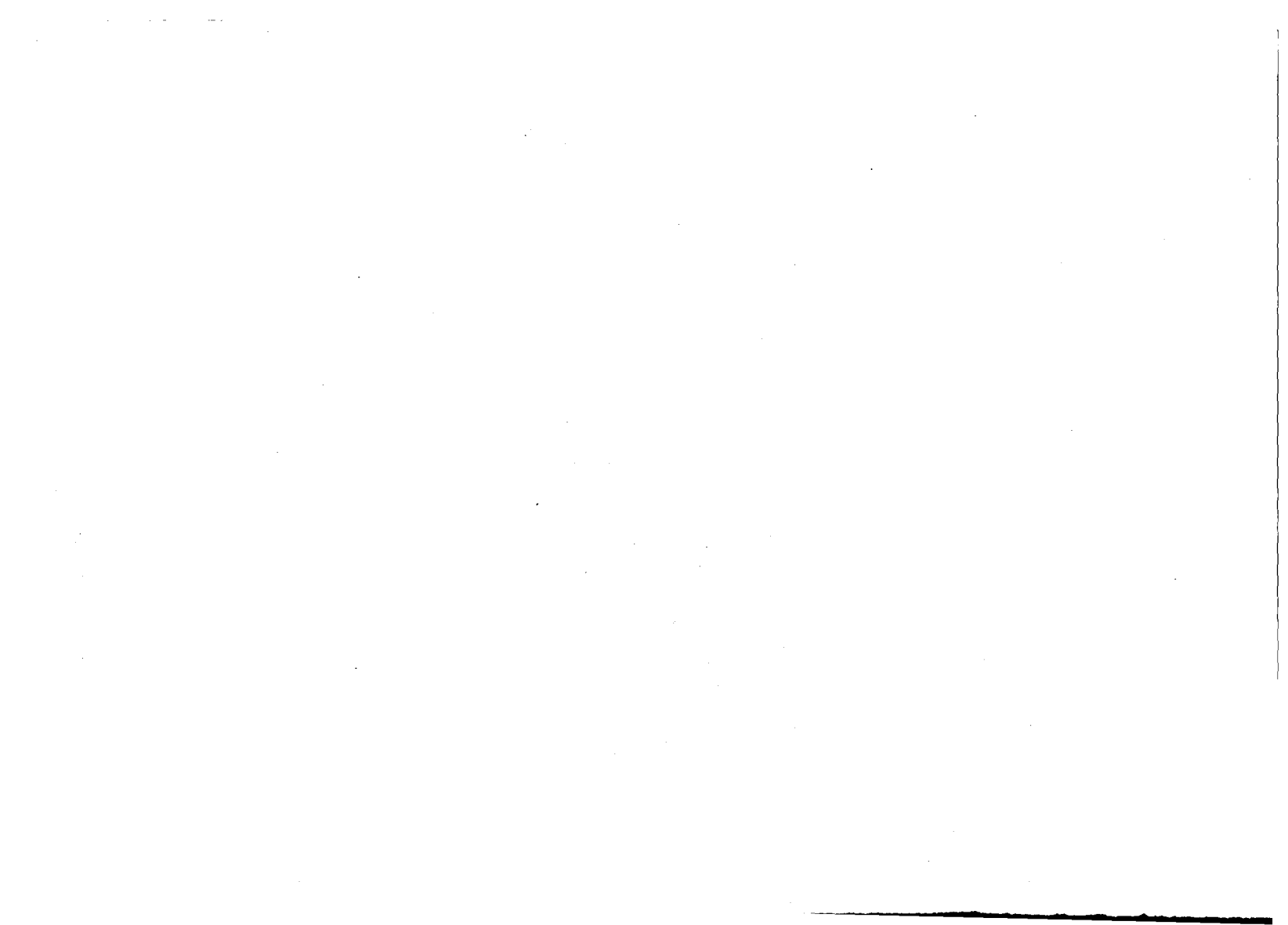
AUTH: A/EXTON, R. J.; B/HILLARD, M. E.

CORP: National Aeronautics and Space Administration. Langley Research Center,
Hampton, Va. AVAIL. NTIS SAP: HC A02/MF A01MAJS: /*DOPPLER EFFECT/*FLOW VELOCITY/*LASER DOPPLER VELOCIMETERS/*MOLECULAR
FLOW/*RAMAN SPECTROSCOPY/*SPECTRAL LINE WIDTHMINS: / FLAT PLATES/ SHOCK WAVES/ STATIC PRESSURE/ SUPERSONIC WIND TUNNELS/
TUNABLE LASERS

ABA: Author

ABS: Molecular flow velocity of N₂ was measured in a supersonic wind tunnel using inverse Raman spectroscopy. This technique employs the large Doppler shift exhibited by the molecules when the pump and probe laser beams are counter-propagating (backward scattering). A retrometer system is employed to yield a vibration-free optical configuration which has the additional advantage of obtaining both the forward and backward scattered spectra simultaneously. The linebreadths and their relative Doppler shift can be used to determine the static pressure, translational temperature, and molecular flow velocity. A demonstration of the concept was performed in a supersonic wind tunnel and included: (1) measurements over the Mach number range 2.50 to 4.63; (2) static pressure measurements (at Mach 2.50)

ENTER:



MOLECULAR VELOCIMETRY USING STIMULATED RAMAN SPECTROSCOPY

R. J. Exton and M. E. Hillard
NASA LaRC
Hampton, VA 23665

ABSTRACT

Molecular flow velocity of N_2 is measured in a supersonic wind tunnel using Inverse Raman Spectroscopy. This technique employs the large Doppler shift exhibited by the molecules when the pump and probe laser beams are counter-propagating (backward scattering). A retrometer system is employed to yield a vibration-free optical configuration which has the additional advantage of obtaining both the forward and backward scattered spectra simultaneously. The linebreadths and their relative Doppler shift can be used to determine the static pressure, translational temperature, and molecular flow velocity. A demonstration of the concept was performed in a supersonic wind tunnel and included (1) measurements over the Mach number range 2.50 - 4.63, (2) static pressure measurements (at Mach 2.50) corresponding to a Reynolds number per foot range of $1 - 5 \times 10^6$, and (3) measurements behind the shock wave of a flat plate model.

N84-25433#

BACKGROUND

Laser Doppler Velocimetry (LDV) has been utilized extensively in the past for measuring flow velocities by tracking particles seeded into the flow. The seeding requirement, however, is often undesirable and sometimes difficult (e.g., flames). In aerodynamic research, strong velocity gradients (e.g., shock waves, vortex flows) make it difficult for the particle to track the flow due to the large inertia of the particle relative to the molecules. The resulting particle lag represents a severe limitation to the application of LDV techniques at supersonic speeds. Seeding of particles into specific regions of interest is also very difficult. For example, seeding into the center of a vortex flow is hampered by the centrifugal forces experienced by the particles which render the vortex center void of particles. A new technique has been proposed⁽¹⁾ to measure the flow velocity of the molecules directly using non-intrusive laser techniques. The molecular velocimeter utilizes the Doppler shift experienced by the molecules as revealed in the Raman spectrum. Spontaneous Raman spectroscopy (single laser) could in principle be used for this purpose, but weak signals and inadequate spectrometer resolution preclude such a measurement. Coherent (stimulated) Raman spectroscopy, however, increases the signal by orders of magnitude and has a considerably higher resolution due to the inherently narrow linewidths associated with lasers. The coherent techniques have been demonstrated in laboratory environments using Stimulated Raman Gain Spectroscopy (SRGS)⁽²⁾, Coherent Anti-Stokes Raman Spectroscopy (CARS)⁽³⁾, and Inverse Raman Spectroscopy (IRS)⁽⁴⁾. In this paper, we will describe some preliminary results of an application of IRS to a major supersonic wind tunnel at Langley Research Center. In order to make these sophisticated laser measurements in a high vibration environment, a retrometer was also developed which reduces the normally double-ended system (two optical tables) to a single optical table plus

retrometer and results in a vibration-free configuration. The retrometer offers an additional advantage by providing simultaneous measurements of both forward and backward scattering. Spectral line shape analysis of these two laser configurations allows temperature and pressure to be measured in addition to velocity.

THEORY OF THE STIMULATED RAMAN TECHNIQUE

In this section, we will describe the theory of coherent (stimulated) Raman spectroscopy as it applies to IRS (and SRGS). In these coherent versions, two focused laser beams interact simultaneously with the molecules and the difference in frequency between them is adjusted to be in resonance with a Raman molecular frequency, Δ . For molecules moving with a velocity V , the (IRS) Raman resonance occurs when

$$\Omega = (\omega - \Delta) + (\bar{k}_{\Omega} - \bar{k}_{\omega}) \cdot V \quad (1)$$

where Ω and \bar{k}_{Ω} are the frequency and wave vector of the pump beam (usually a pulsed laser) and ω and \bar{k}_{ω} are the frequency and wave vector of the probe beam (usually a C.W. laser). Both laser beams are tightly focused at the same point in space and one of them is tunable in order to come into resonance with the moving molecules. Many geometrical arrangements can be envisioned for these two lasers; however, two configurations define the limits of their interactions with the molecules - namely, forward and backward scattering. In forward scattering, both laser beams co-propagate from the same direction; whereas in backward scattering, the laser beams are in a counter-propagating configuration. In the forward scattering case, moving molecules "see" two laser beams whose frequencies are shifted in the same (spectral) direction and by about the same amount; hence there is very little Doppler shift (and breadth) by which to measure velocity. In the backward scattering case, on the other hand, the molecules "see" one laser beam shifted in one spectral direction, while the other laser beam is shifted in the opposite direction. This results in a large Doppler shift (and

breadth) which is inherently easier to measure. The stimulated Raman interaction takes place over a very small volume of space defined by the overlap and focused character of the beams. In the quasi-C.W. arrangement⁽⁵⁾ discussed here, the interaction is sensed as a gain (SRGS) or loss (IRS) in the probe laser power only during the brief time (typically 5-10 nano-seconds) that the pump beam is present in the interaction volume. The probe signal loss $\delta P(\omega)$ for IRS corresponding to two focused, collinear, Gaussian beams is given by^(5,6)

$$\delta P(\omega) = \frac{4 \cdot 10^7 \pi^4 P(\omega) P(\Omega) \omega \Delta N (d\sigma/d\Omega)}{hc^2 n^2 \Delta\omega k_\Omega^3} \quad (2)$$

where

- $P(\omega)$ - probe laser power (W)
- $P(\Omega)$ - pump laser power (W)
- ΔN - population difference between the lower and upper states (cm^{-3})
- $d\sigma/d\Omega$ - differential Raman scattering cross section ($\text{cm}^2 \text{sr}^{-1}$)
- n - index of refraction at the pump frequency
- $\Delta\omega$ - Raman linewidth (sec^{-1})

The difference between the forward and backward signals in equation (2) lies entirely with the Raman linewidth $\Delta\omega$. The major contributions* to this linewidth are Lorentz (pressure) broadening [$\Delta\omega_L = K P(1/T)^{1/2}$] which is the same for both forward and backward scattering and the Doppler breadth, $\Delta\omega_D$,

*Dicke narrowing may slightly modify the breadths in the region near 0.2 amagat⁽⁷⁾, but there has not yet been a systematic investigation of diffusional effects for N_2 , the molecule of prime importance here. Optical Stark effects must also be considered and set an upper limit on the practical pump laser intensity that can be used.

given by

$$\Delta\omega_D = \left(\frac{8kT \ln 2}{mc^2} \right)^{1/2} (\omega \pm \Omega) \quad (3)$$

for forward (-) and backward (+) scattering. The resultant Voigt breadths for forward and backward scattering are, therefore, determined by the gas temperature and pressure. Figure 1 illustrates these effects for a line in the Q branch of N₂ at 100°K. Inversely, the forward and backward breadths jointly determine the temperature and pressure (Fig. 2). Simultaneous measurements of both forward and backward line shapes for a moving gas would render a result similar to that sketched in Figure 3. The difference in frequency shifts between the forward and backward IRS peaks is given by $W = 2\omega V/C \cos \theta$ where θ is the angle between the (collinear) laser directions and the velocity vector. In this manner, velocity can be measured without resorting to an absolute frequency measurement. All three parameters; velocity, pressure, and temperature (translational), can be determined from frequency difference measurements independent of intensity or absolute frequency considerations.

EXPERIMENTAL APPARATUS

A schematic of the optical configuration is shown in Fig. 4 in which the focus point (interaction volume) is centered in the test section of a wind tunnel. The pump laser is the output of a pulsed dye amplifier (PDA) which is used to amplify the tunable, narrowband (1 MHz) C.W. dye laser input. The PDA is pumped by the Q-switched pulses from a frequency doubled YAG laser. The resultant tunable output of the PDA typically has a bandwidth of 190 MHz and 7 mJ/pulse at 5846Å. The probe laser is a single longitudinal mode (linewidth <10 MHz) Ar⁺ laser operated at 5145Å with power output typically 500 mW. The Ar⁺ probe and PDA pump beams are then combined through two Pellin-Broca prisms and focused by lens L₁ into the sample volume (approximately 200 μm diameter by 6 cm

long). Optical losses attenuate the laser beams incident on the sample volume to approximately 5 mJ/pulse at 5846Å and 240 mW at 5145Å. The two focused beams then intersect lens L_2 (identical to L_1) which renders two (collinear) collimated beams. The pump beam is dumped at the retrometer using a dichroic beam splitter and a light trap. The collimated probe beam is transmitted by the beam splitter and allowed to strike the corner cube whereby this retro-reflected beam is brought to the same focus point in the sample volume as the forward beams. This situation is obtained since a corner cube sends light back in the same direction from whence it came. The result is that this retro-reflected focus point is practically independent of lateral or rotational movements of the retrometer. In this manner, counter-propagating, focused beams are easily attained with the simple requirement that the forward probe beam strike the retrometer. In this configuration, the retrometer can be moved (shaken) relative to the laser table, but this motion will not perceptively impact the overlapping of the two counter-propagating beams. Vibration-free overlap of the two counter-propagating beams is, therefore, attained. The retro-reflected probe beam returns to the optical table where an optical isolator (Faraday rotator) intercepts the beam and directs it to a silicon photodiode detector (incident probe power typically 35 mW). The retro-reflected probe beam is then analyzed with a two channel boxcar integrator which measures the change (gain or loss) in the probe beam that occurs during the pulse duration of the pump laser.

In the configuration shown in Fig. 4, the pump beam (directed toward the retrometer) simultaneously interacts with the C.W. probe beam in both the forward and backward directions. For a given pulse, the detector will first see a signal (gain/loss) from the backward scattering case, followed by a signal from the forward scattering case. This latter signal will be observed at a later time given by the round-trip time for the probe beam to travel from the sample volume (focus point) to the retrometer and back. This delay for the present

configuration is about 10 nsec - easily discriminated by the two channels in the boxcar integrator. Thus by scanning the PDA, forward and backward spectra are observed simultaneously.

MEASUREMENTS IN A SUPERSONIC WIND TUNNEL

In order to demonstrate the molecular velocimetry technique and the present system's capability, we have conducted tests in the high Mach number test section of the Unitary Plan wind tunnel at the Langley Research Center. In this test section, continuous air flow can be reliably established over the Mach number range 2.29 - 4.63 and a Reynolds number per foot (Re) range of 0.76×10^6 - 7.78×10^6 . The non-intrusive laser tests consisted of three parts; (1) measurements at five Mach number conditions in order to check velocity and temperature measurements, (2) measurements over a range of Reynolds number (at a fixed Mach number) in order to systematically demonstrate the system sensitivity to static pressure and (3) measurements behind the attached shock of a 2-D flat plate model in order to assess beam steering effects in traversing a shock layer. Many of the tests in (1) and (2) were conducted in the free-stream region in front of research aircraft models in order to conserve tunnel operating time and expense. An expanded plot of the forward and backward linebreadths expected for the free stream flows in the Unitary Plan wind tunnel is shown in figure 5. The linebreadths computed for this plot include the PDA linebreadth (190 MHz) in order to render pressure and temperature directly from the measured spectral data. A sample data point is shown in figure 5 corresponding to the spectral data shown in figure 6 for a Mach 2.50 free stream flow ($Re = 2 \times 10^6$). The measured forward and backward linebreadths and shift for this point are $\Delta\nu_F = 0.33$ GHz, $\Delta\nu_B = 1.98$ GHz, and $W_\nu = 1.00$ GHz, in linear frequency, respectively. The breadths correspond to a pressure of 35 torr at a temperature of 161°K. The shift corresponds to a velocity of 258 m/sec along

the laser beam direction. Since the laser beams were inclined 64.6° with respect to the flow direction, this translates into a flow velocity of 602 m/sec (Mach number = 2.37). One of the contributions to the noise shown in figure 6 is due to laser fluctuations since no attempts thus far have been made to normalize the signals. The averaging provided by the boxcar integrator, however, is adequate to demonstrate the concepts. For example, figure 7 shows the measured Mach number plotted versus the aerodynamically set conditions. The Mach number is the ratio between the flow velocity and the local speed of sound. Since the speed of sound is a function of the local temperature, the measured Mach number reflects both the measured velocity and measured static temperature. The aerodynamically set Mach number is accurate to 0.02. The data points in figure 7 represent the mean of the measured Mach numbers at each set condition while the bars show the standard deviation about the mean. The mean measured Mach number is in good agreement with the set conditions, showing a tendency to be low by about 5%. The major contribution to the random error here is the determination of temperature which is influenced primarily by the backward breadth. Although this breadth is dominated by the Doppler contribution, the backward signal is significantly weaker than the forward signal due to the increased breadth* (refer to eqn. 2).

The capability for measuring static pressure is illustrated in figure 8. For these tests, the Mach number was held constant at 2.96 while the static pressure was set at five different levels corresponding to $Re = 1, 2, 3, 4$, and 5×10^6 . The aerodynamically set static pressures were accurate to approximately 1%. The data in figure 8 represent individual measurements of static

*Measured peak signal levels, $\delta P(\omega)/P(\omega)$, for the wind tunnel experiments were typically 1×10^{-3} for forward scattering and 2×10^{-4} for backward scattering.

pressure. These data illustrate the capability of the technique for measuring pressure, but show a consistent trend toward higher values. A large part of the deviation would appear to be caused by a constant offset of the data, best exemplified by the linear least squares fit. Parameters in the model which could account for deviations in measured pressure include (1) the validity of the assumed $T^{-1/2}$ dependence for the Lorentz broadening and (2) changes in the PDA breadth since calibration. The PDA breadth of 190 MHz is particularly important at the lower pressures since it represents a sizeable contribution to the measured forward linebreadth which is dominated by the Lorentz (pressure) contribution (refer to figure 1).

The shock layer tests were performed with a flat plate model 45 x 45 x 2 cm (see Fig. 9). This large 2-D model was chosen since the conditions behind the shock are uniform over appreciable spatial dimensions and can also be accurately predicted. The model was moveable in the X (flow) direction which allowed the IRS interaction volume to be placed at different locations with respect to the shock wave. The probe laser beam traverses the shock wave four times when making measurements behind the shock--twice at the side of the model nearest the optical table and twice near the leading edge. No deleterious effects due to beam steering were observed in the IRS signal for the full range of wind tunnel conditions encountered. A comparison of measurements made before and after the shock with calculated values are shown in Table I for a Mach 2.96 flow. The measured values compare well with the calculated ones with the possible exception of the temperature before the shock. The static pressures measured here are also high following the trend established earlier (see figure 8). In this regard, it should be noted that the ratio of these quantities (after/before shock) agree more closely with the calculated ratios, implying that the measurements are internally consistent, although not as accurate as desired.

CONCLUSIONS

Molecular flow velocity, static pressure, and translational temperature have been simultaneously measured in a supersonic wind tunnel using stimulated Raman spectroscopy. This represents the first time that all of these critical aerodynamic quantities have been measured using a single non-intrusive measurement technique. The additional significance of these measurements are (1) that the pressure is measured directly rather than being inferred from a density (and temperature) measurement, (2) that the translational temperature is measured--not a vibrational or rotational temperature, and (3) that the velocity of the major molecular constituent, N_2 , is measured--not inferred from other seeded species or particles. The technique was rendered practical in the high vibration tunnel environment through the use of a specially designed retrometer. The retrometer also allows simultaneous measurements of both the forward and backward scattered Raman lines. Analysis of the breadths and relative Doppler shift of these lines renders a single data point for velocity, pressure, and temperature. Mach number comparisons with aerodynamic set conditions showed good agreement demonstrating the measurement capability for velocity and temperature. Static pressure also agreed well with aerodynamic set conditions over a range of Reynolds number. Although we are not concerned at this time with a detailed discussion of the measurement precision, these measurements adequately demonstrate the fundamental concepts involved.

The demonstrated ability to perform measurements behind a shock layer are also significant and opens the way for detailed studies of complex aerodynamic phenomena. Increased spatial resolution, measurement precision, and instrument response time are factors which will see significant improvements in the near future. In addition, multi-component analysis of the velocity vector through the use of multiple beams and intermediate geometries will certainly be a central theme of continued research.

REFERENCES

1. She, C. Y.; Fairbank, W. M., Jr.; and Exton, R. J.: Measuring Molecular Flows with High-Resolution Stimulated Raman Spectroscopy. IEEE J. Quant. Electr. QE-17, 2-4 (1981).
2. Herring, G. C.; Fairbank, W. M., Jr.; and She, C. Y.: Observation and Measurement of Molecular Flow Using Stimulated Raman Gain Spectroscopy. IEEE J. Quant. Electr. QE-17, 1975-76 (1981).
3. Gustafson, E. K.; McDaniel, J. C.; and Byer, R. L.: CARS Measurement of Velocity in a Supersonic Jet. IEEE J. Quant. Electr. QE-17, 2258-59 (1981).
4. Herring, G. C.; Lee, S. A.; and She, C. Y.: Measurements of a Supersonic Velocity in a Nitrogen Flow Using Inverse Raman Spectroscopy. Opt. Lett. 8, 214 (1983).
5. Esherick, P.; and Owyong, A.: "High-Resolution Stimulated Raman Spectroscopy" in Advances in Infrared and Raman Spectroscopy, Vol. 9, Clark, R. J. H. and Hester, R. E., Eds. Heyden and Son, Ltd., 1982, pp. 130-187.
6. Yeung, E. S.: "Applications of Inverse Raman Spectroscopy" in Chemical Applications of Nonlinear Raman Spectroscopy, Harvey, A. B., Ed., Academic Press, 1981, pp. 171-204.
7. Rosasco, G. J.; Lempert, W.; Hurst, W. S.; and Fein, A.: "Pressure Dependent Linewidth and Line Shift Measurements in the Vibrational Q-Branch of N₂ from 4 to 200 kPa" in Spectral Line Shapes, Vol. 2, Proceedings of 6th Int. Conf. on Spectral Line Shapes, Burnett, K., Ed. Walter de Gruyter, 1983, pp. 635 - 649.

Table I: FLAT PLATE MODEL--Measurements before
and after shock ($M = 2.96$, $\alpha = 15.6^\circ$, $\phi_s = 33.3^\circ$)

	Before Shock		After Shock		Ratio (After/Before)	
	<u>Calc.</u>	<u>Meas.</u>	<u>Calc.</u>	<u>Meas.</u>	<u>Calc.</u>	<u>Meas.</u>
V (m/sec)	644	614	564	531	0.88	0.86
T ($^\circ\text{K}$)	118	134	165	167	1.40	1.25
P (torr)	21	23	62	70	2.95	3.04
Mach #	2.96	2.67	2.19	2.03	0.74	0.76

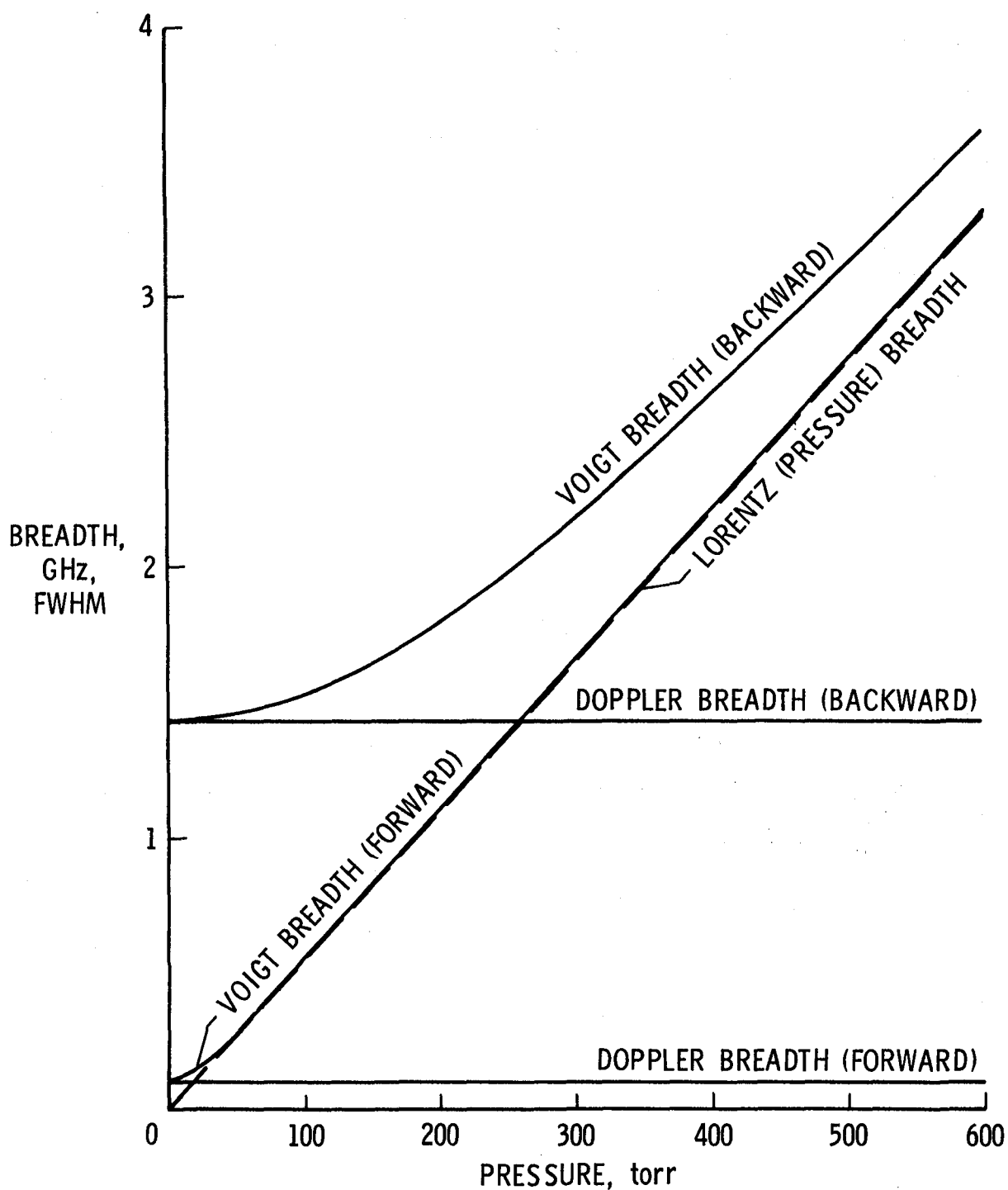


Fig. 1 Backward and forward linebreadths (FWHM) are shown as a function of pressure for the N_2 Q branch ($J = 10$) line at $100^\circ K$.

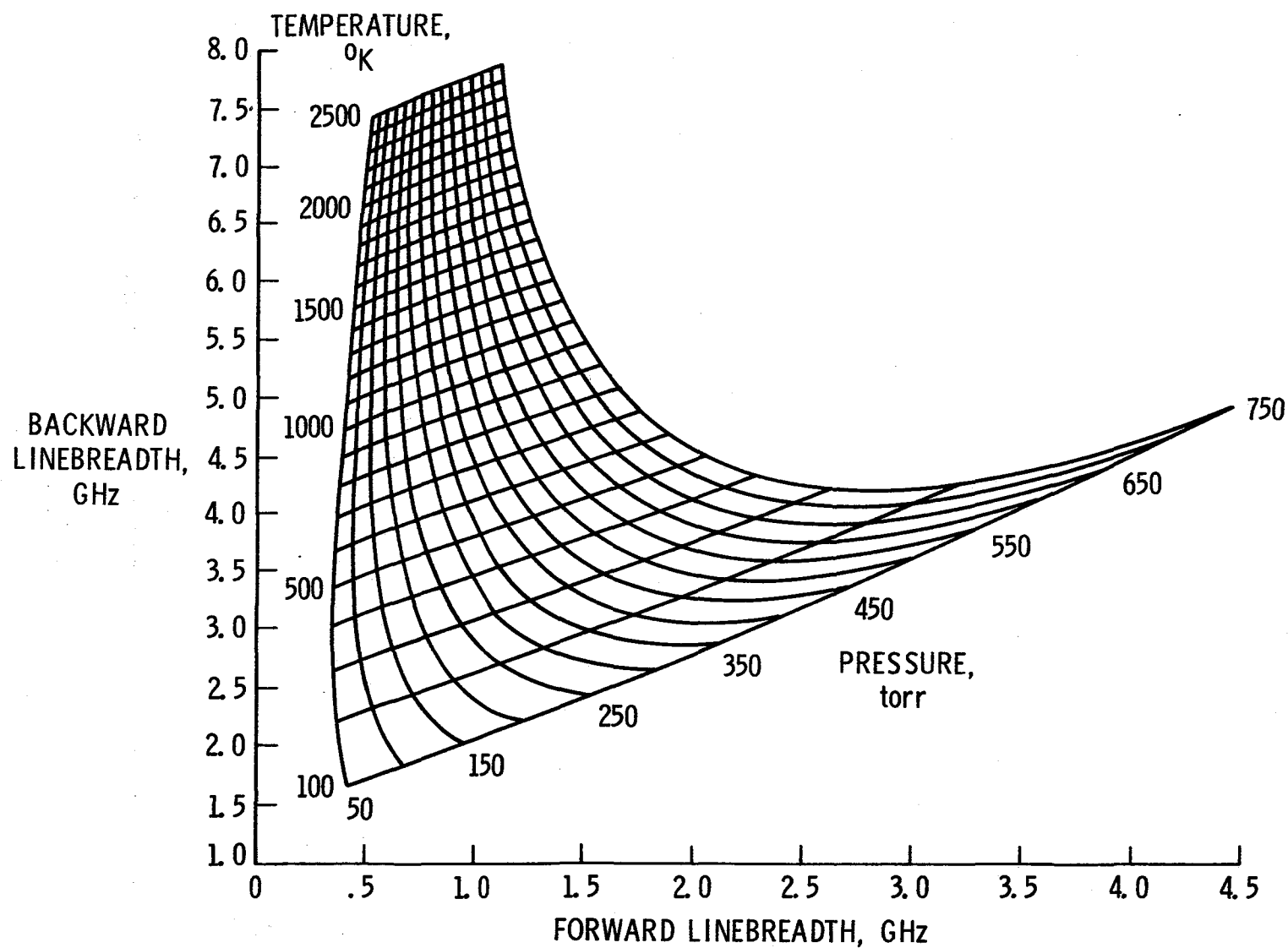


Fig. 2 Backward and forward Voigt linebreadths (FWHM) are shown for the N_2 Q Branch ($J = 10$) line over a wide range of temperature and pressure.

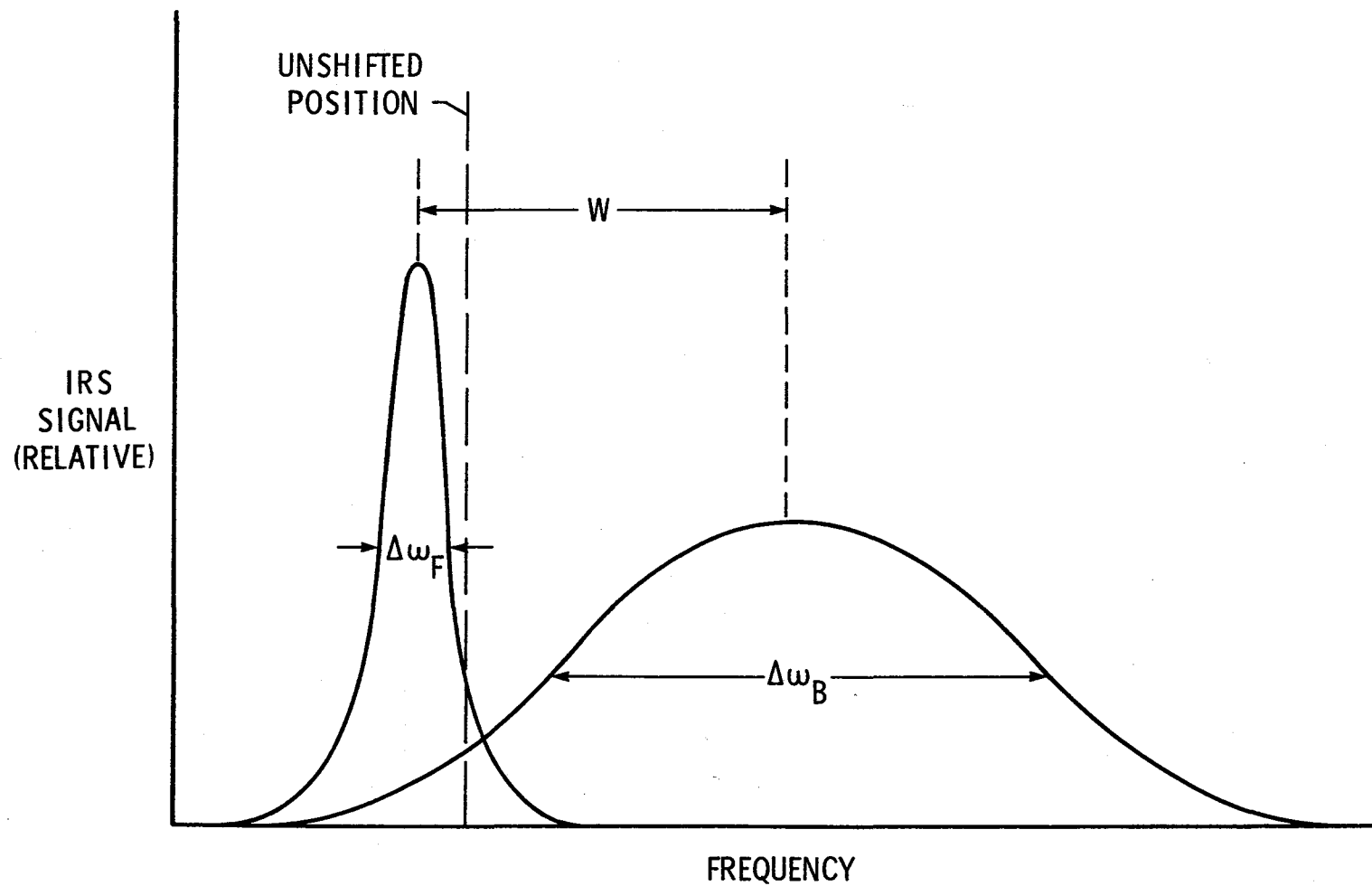


Fig. 3 Sketch of the shift and breadth of the forward and backward scattered Raman lines relative to the unshifted (zero velocity) position.

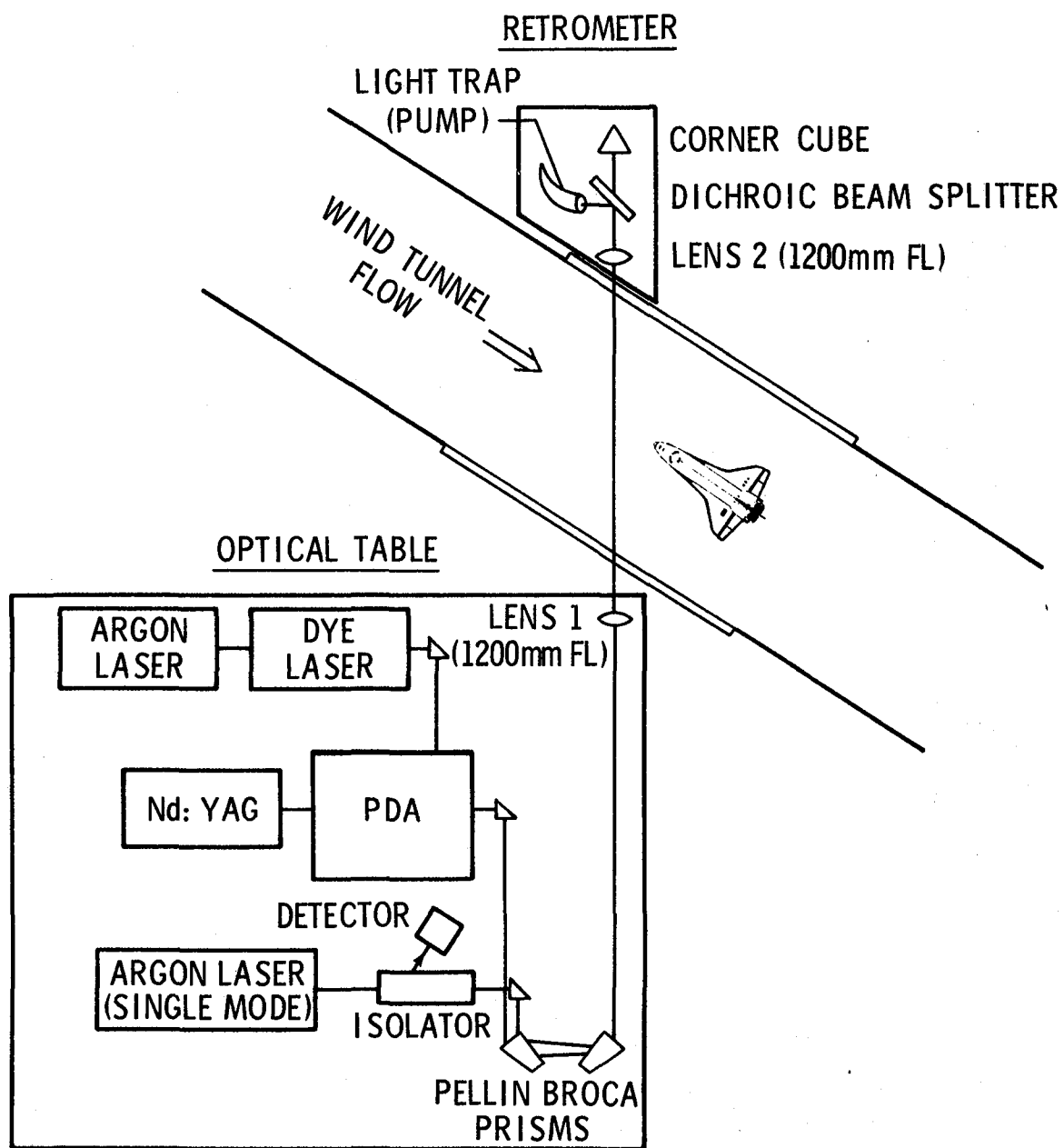


Fig. 4 Schematic of the optical configuration for vibration-free Raman Doppler velocimetry in a wind tunnel.

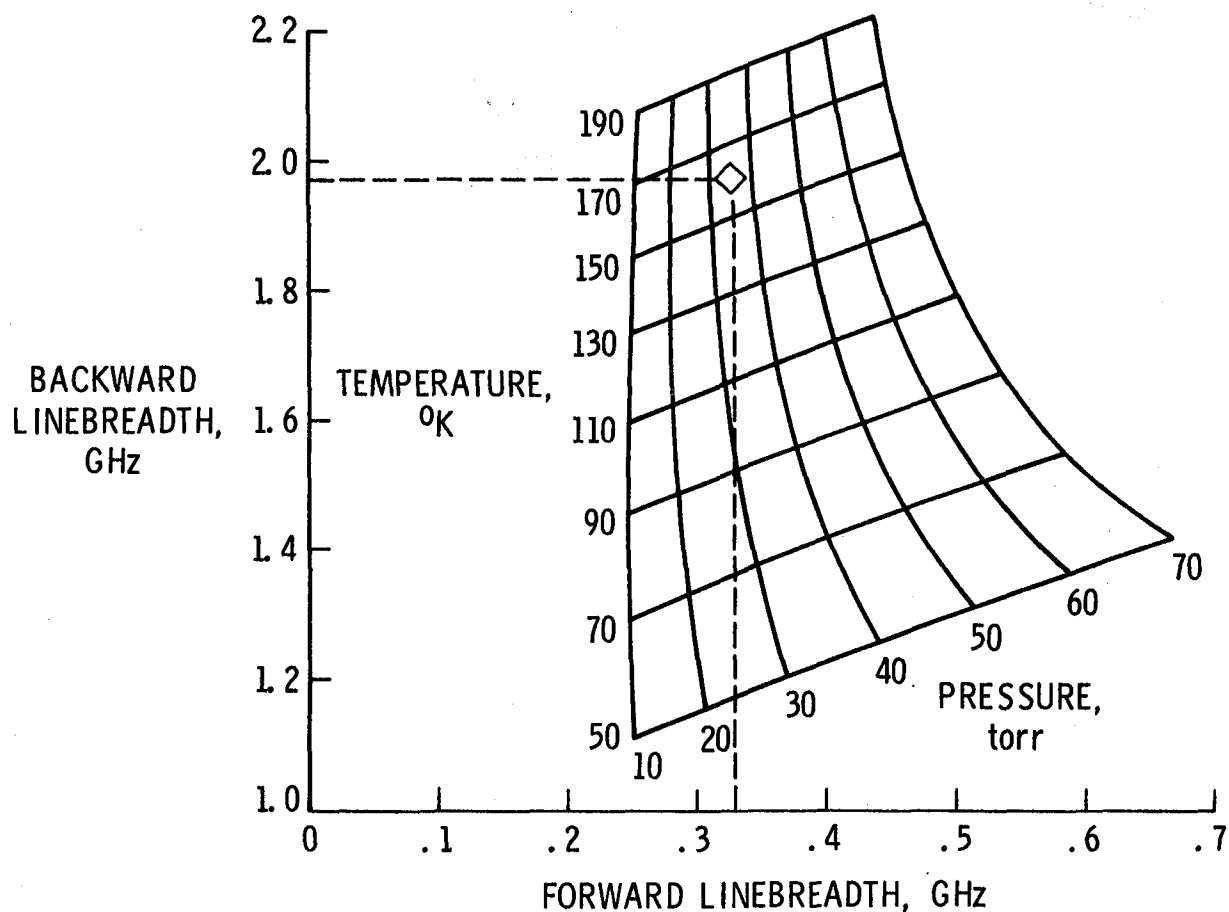


Fig. 5 Backward and forward linebreadths for the N₂ Q branch (J = 10) line for the temperature and pressure range expected for supersonic free stream flows in the Langley Unitary Wind Tunnel. The 190 MHz instrumental linewidth (assumed Gaussian) of the PDA is included in the Voigt breadths (FWHM) shown here. A single data point is also shown corresponding to the spectral data shown in figure 6.

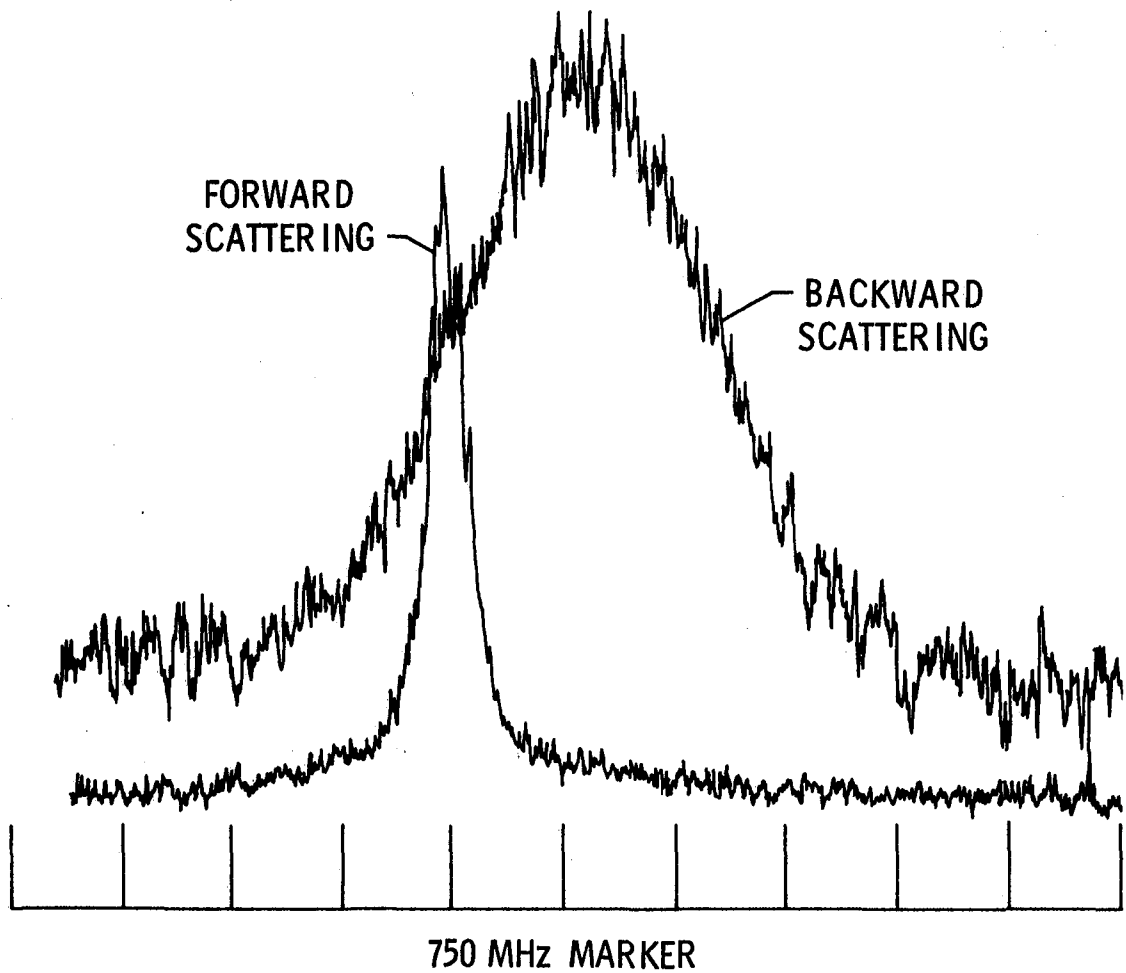


Fig. 6 Measured spectral data for a Mach 2.50 free-stream flow in the Unitary Wind Tunnel ($Re = 2 \times 10^6$). The scan time was approximately 2 minutes.

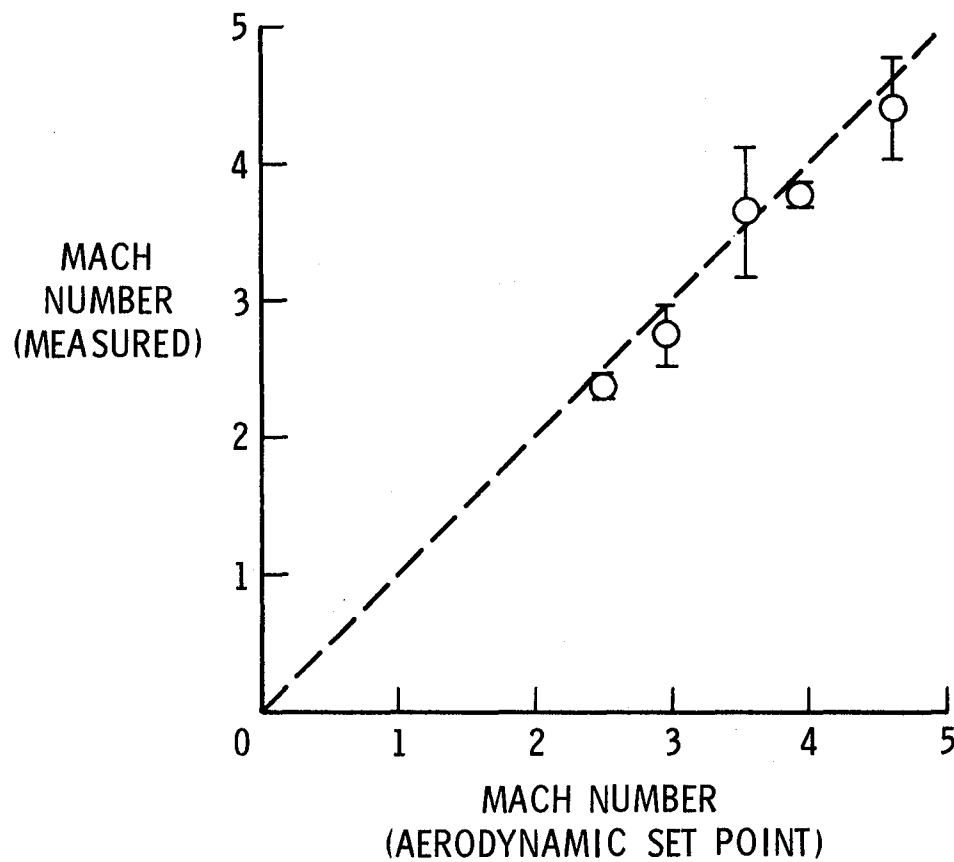


Fig. 7 Measured free-stream Mach number versus the aerodynamically set Mach number in the Unitary wind tunnel ($Re = 2 \times 10^6$ for all points except M4.63 for which $Re = 4 \times 10^6$)

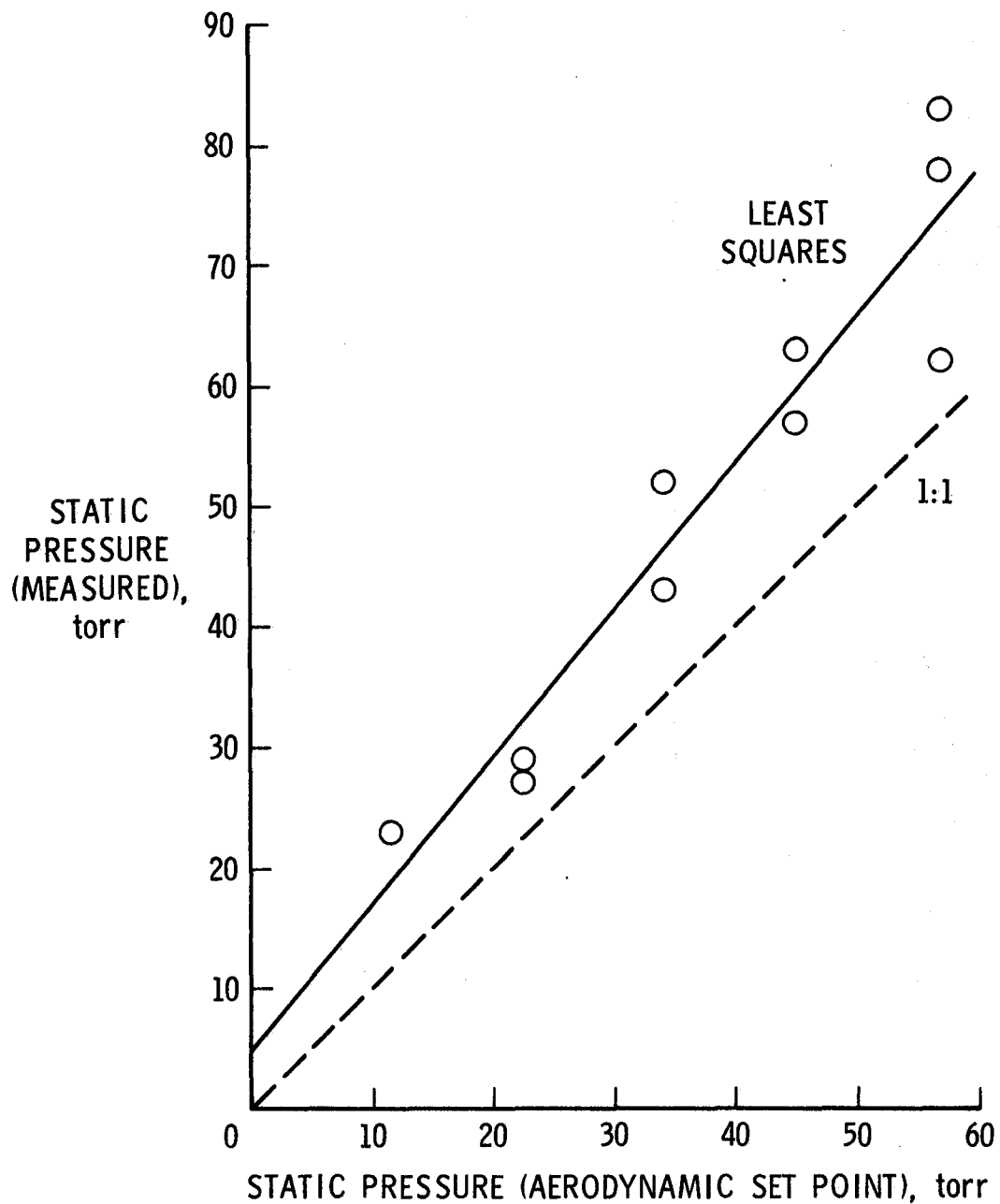


Fig. 8 Measured static pressure versus aerodynamically set pressures corresponding to $Re = 1, 2, 3, 4$, and 5×10^6 . The mean of the measured temperatures for these points was $121^\circ K$ (standard deviation $17^\circ K$) compared with a computed temperature of $123^\circ K$.

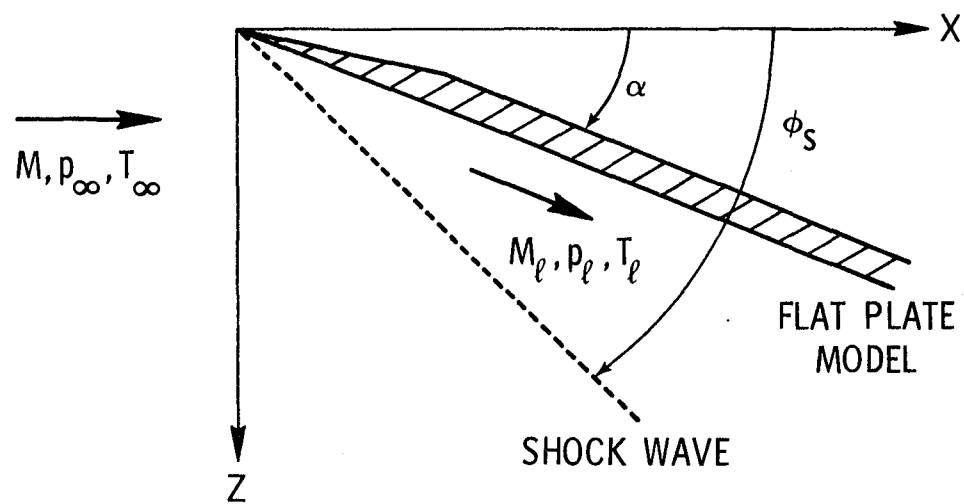


Fig. 9 Cross-sectional sketch of the flat plate model and its attached shock wave

1. Report No. NASA TM-85808		2. Government Accession No.		3. Recipient's Catalog No.	
4. Title and Subtitle Molecular Velocimetry Using Stimulated Raman Spectroscopy				5. Report Date May 1984	
				6. Performing Organization Code 505-31-53-13	
7. Author(s) Reginald J. Exton and Mervin E. Hillard				8. Performing Organization Report No.	
9. Performing Organization Name and Address NASA Langley Research Center Hampton, VA 23665				10. Work Unit No.	
				11. Contract or Grant No.	
12. Sponsoring Agency Name and Address National Aeronautics and Space Administration Washington, DC 20546				13. Type of Report and Period Covered Technical Memorandum	
				14. Sponsoring Agency Code	
15. Supplementary Notes					
16. Abstract Molecular flow velocity of N ₂ is measured in a supersonic wind tunnel using Inverse Raman Spectroscopy. This technique employs the large Doppler shift exhibited by the molecules when the pump and probe laser beams are counter-propagating (backward scattering). A retrometer system is employed to yield a vibration-free optical configuration which has the additional advantage of obtaining both the forward and backward scattered spectra simultaneously. The linebreadths and their relative Doppler shift can be used to determine the static pressure, translational temperature, and molecular flow velocity. A demonstration of the concept was performed in a supersonic wind tunnel and included (1) measurements over the Mach number range 2.50 - 4.63, (2) static pressure measurements (at Mach 2.50) corresponding to a Reynolds number per foot range of 1 - 5 x 10 ⁶ , and (3) measurements behind the shock wave of a flat plate model.					
17. Key Words (Suggested by Author(s)) Velocimetry Raman Stimulated Raman Doppler			18. Distribution Statement Unclassified - Unlimited Subject Category - 72		
19. Security Classif. (of this report) Unclassified		20. Security Classif. (of this page) Unclassified		21. No. of Pages 22	
				22. Price A02	

